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## Physische eigenschappen van bolvormige moleculen

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## SUMMARY

In this thesis the properties of spherical molecules have been investigated, especially of molecules, belonging to the type  $C\alpha_4$ . These compounds show a number of remarkable properties, which are to be explained by the supposition, that the molecules possess a certain freedom of orientation in the lattice.

In the present investigation work is done on the question, how this freedom of orientation should be interpreted: either in the sense of an isotropic rotation or in the sense of a statistic distribution of the molecules over a number of different, but energetical equivalent positions in the lattice.

From the compounds prepared in the present laboratory (list p. 9), the most characteristic representative was selected by means of physicochemical methods, viz. the tetramethylorthothiocarbonate  $C(SCH_3)_4$ .

This compound shows three modifications:

Modification I, stable below  $23.2^\circ C$ ; modification II, stable between  $23.2$  and  $45.5^\circ C$  and modification III (cubic), stable from  $45.5^\circ C$  up to the melting point  $65.7^\circ C$ .

The main results of the X-ray investigation are as follows:

Modification I. Space group  $D_{2d}^4$ ; the crystals are piezo-electric. The unit cell contains two molecules, one in the angular points of the cell, the other in the center. The mutual position of the two

molecules is such, that the symmetry operations of the glide plane (110) with a component  $\frac{1}{2}(a + b + c)$  convey them into each other (fig. 23). The smallest distance between the molecules is found in the direction of the  $c$ -axis: the methyl groups on both sides of this axis touch each other (fig. 27 I).

Modification II. Space group  $D_{4h}^{17}$ ; the crystals do not show piezo-electricity. The lattice is body-centred; by considering the spatial relations it is seen, however, that the centre of symmetry as well as the body-centring of the separate unit cell cannot be real. Both phenomena are simulated to us by the co-operation of a number of unit cells, for the molecule  $C(SCH_3)_4$  fits into the lattice in two positions ( $a$ ) and ( $b$ ). If one imagines a molecule in a fixed position in the corner of the unit cell, one can find the position ( $a$ ) of another molecule in the centre by an inversion with regard to the point  $(\frac{1}{4} \frac{1}{4} \frac{1}{4})$ , and the position ( $b$ ) by the operation of the glide plane (110) with a component  $\frac{1}{2}(a + b + c)$ . Moreover the methyl group can occupy two positions with respect to the tetrahedron, formed by the sulphur atoms. The crystal shows the symmetry of the classe  $D_{4h}$ , because all these positions are superposed (fig. 27 III).

Modification III (cubic). The space group is  $O_h^9$ . The unit cell is body-centred. The symmetry of the molecule can be brought in agreement with the high point symmetry of its position in the lattice, if it is assumed, that the methyl group can occupy three positions with regard to the sulphur-tetrahedron (fig. 27 III). An isotropic rotation can be excluded completely because of lack of space.

Finally the properties of the spherical molecules have been treated theoretically and interpreted in a statistical and thermodynamical way. As a description of the remarkable cubic phase of the spherical molecules, the term „dynamical crystal” has been proposed.